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# Time-resolved spectroscopic investigation of emission observed during damage in the bulk of fused silica and DKDP crystals

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## ABSTRACT

We have investigated the flash of light that accompanies laser damage using time-resolved spectroscopy. Damage events were initiated in the bulk of both fused silica and DKDP crystals with 355-nm 3-ns pulsed radiation. Spectra from the accompanying flash were recorded in the 200 - 500 nm wavelength range with 5-ns temporal resolution. Ten ns following damage initiation, the spectra were found to be roughly blackbody with temperatures on the order of 5000 K to 7000 K, depending on the material studied and excitation energy used. The observed temperatures and cooling rates can be related to the size and electron density of the plasma "fireball" that initiates the damage event.

**Keywords**: fused silica. DKDP crystals. spectroscopic investigaion

## INTRODUCTION

The observed laser damage threshold in optical materials is significantly less than the theoretical intrinsic value. In fused silica, for 350-nm light, it has been estimated that the onset of avalanche breakdown should be on the order of 150 GW/cm$^2$.[1] For the 3-ns pulses used in this study, this corresponds to 450 J/cm$^2$, which is 40 to 50 times the observed values. This discrepancy is generally attributed to the presence of defects, intrinsic or extrinsic, contained within nominally transparent materials.[2,3]

When laser damage occurs, plasma is formed at the sites of these defects and rapidly expands, absorbing large amounts of energy during the laser pulse.[4,5] The high energy density produces a micro-explosion, damaging the material. Shockwave propagation induces mechanical failure and material modification. This is accompanied on a slower time scale by melting and further material modification caused by the temperature increase.[6,7] The white light emissions, which accompany these processes, contain crucial information concerning the mechanisms responsible for the damage. In this work, we report that the emission spectrum of both DKDP and fused silica are blackbody in nature as early as 10 ns after the termination of the laser pulse and that the ionized region can approach temperatures of 7000 K.

## EXPERIMENTAL PROCEDURE

Fused silica ($SiO_2$) and DKDP ($KH_{2-x}D_xPO_4$) samples at room temperature were individually irradiated with 355-nm 3-ns pulse radiation focused in the bulk of the materials with a spot determined by a knife-edge measurement to be approximately 30 μm in size. The pump beam is brought in through a small turning mirror and focused by an off-axis parabolic mirror into the sample. A schematic representation of the apparatus may be seen in figure 1. The short focal length of the mirror eliminated the possibility of surface damage and allowed the study of the bulk properties of the materials in question.

The same mirror collects the emission from the damage area with an f-number of unity. The collimated refection is transported by reflective optics to an f-4 mirror and focused into a 200-μm spectrometer slit. The 50-μm collection region from this geometry closely matches the spot size of the damaging beam and thus reduces extraneous signal from sources such as bulk fluorescence. By using only reflective optics we prevent excessive loss in UV that would be experienced with refractive optics. The response of the system and absorption of the sample was determined by use of calibrated light sources.

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The emission is dispersed spectrally and gated temporally by a spectrometer/ICCD combination. The spectral and temporal resolutions of the system are 5 nm and 5 ns, respectively.

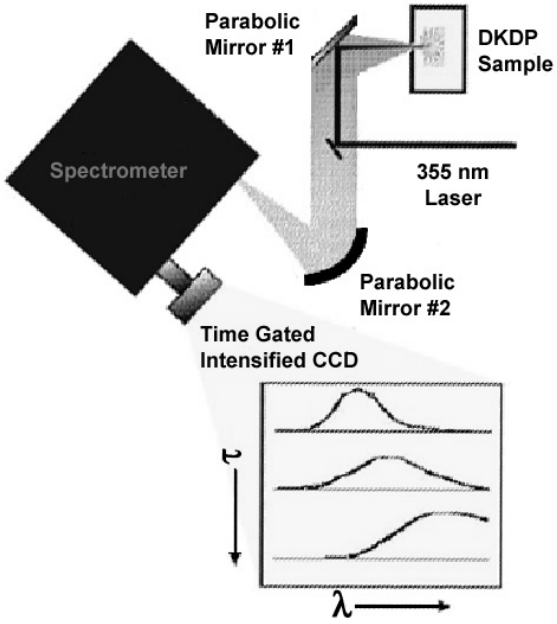


Figure 1

The 355 nm light is reflected off a small turning mirror and then focused by an off-axis parabolic mirror in to the bulk of the sample. After the same mirror collects the light, the collimated beam passes the turning mirror with about 10% loss. A second mirror focuses the light into a spectrometer coupled with a time gated intensified CCD.

To prevent damage to the ICCD by scattered laser light, a physical block was used to obscure the ICCD pixels for 50 nm around either side of the laser line. This is manifested in the data as a gap in the spectrum from ~300-nm to ~400-nm. Emission spectra were obtained for irradiation energies of 600 μJ for delay times in the range of 10 ns to 100 ns for DKDP and 10 ns to 200 ns for fused silica

For DKDP, spectra were collected from virgin (not previously exposed to laser radiation) sites separated by at least 3 times the beam width from one another. No site on the fused silica sample studied was damaged on an initial pulse with the energies used in this study. Instead each location was irradiated an average of 10 times before observable damage occurred. The energy was chosen so that most sites would eventually damage. After damaging, subsequent pulses at a site produced larger signals.

## RESULTS

The data were reduced by removing background and instrument response. Commercial software was then used to fit the spectra to

$$I(\lambda) = C_1 + C_2\lambda^{-5}(e^{0.144/T\lambda} - 1)^{-1} \quad (1)$$

where $C_1$, $C_2$ and T are fitting parameters. The data for DKDP and fused silica are displayed in figures 2a and 2b.

Fitting with a pure Planckian distribution gives a slightly worse fit, but yields temperatures that differ by no more than 10%. This discrepancy does not effect the conclusions of this work.

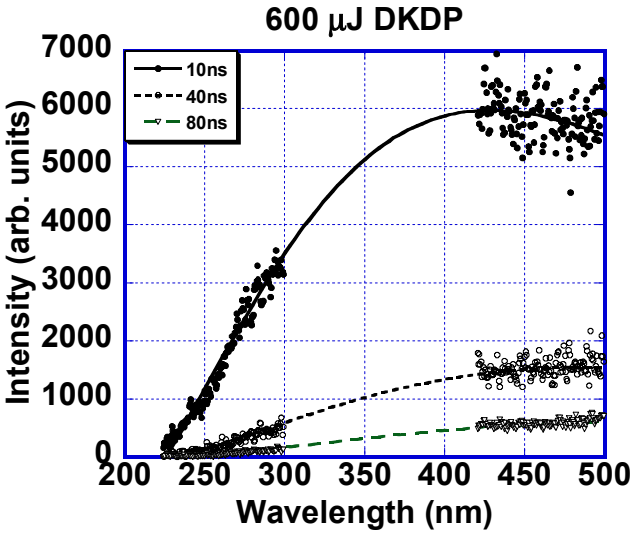


Figure 2a

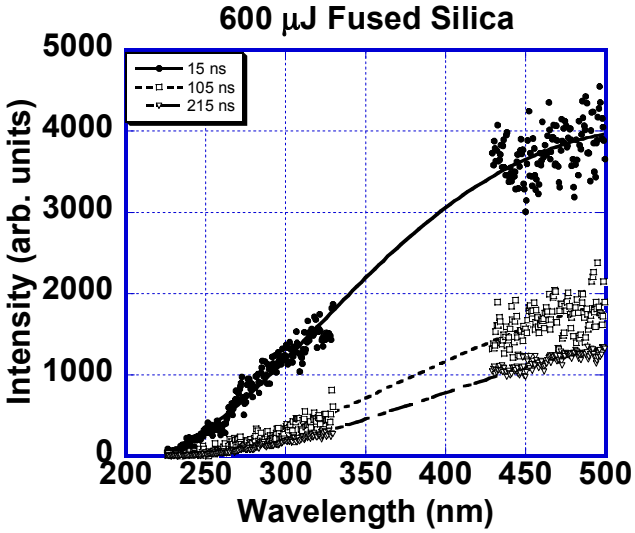


Figure 2b

The individual points are the data collected from the ICCD. The curves through the data are the fits generated by equation 1. Gaps in the spectra are the result of an obstruction blocking the laser line. a. Three of the spectra obtained from DKDP at delay times of 10, 40, and 80 ns. b. Spectra from fused silica for 15, 105, and 215 ns after the laser pulse.

The fit is well constrained by the data on both sides of the peak emission. A spectral window only above 500 nm would miss the peak of the emission at short times and would result in a poorly constrained fit. Without nanosecond-scale resolution the rapid changes in temperatures would make the spectra appear non-thermal.

Extracting the temperature from Equation 1 for each spectrum produced plots of temperature vs. delay time for both materials studied. It can be seen in figure 3 that the materials are at temperatures of approximately 5000 and 7000 K for fused silica and DKDP respectively 10 ns after the laser pulse terminates. One hundred nanoseconds after the pulse the temperature of the DKDP has dropped close to 2000 K while that of the fused silica is only about 500 K lower.

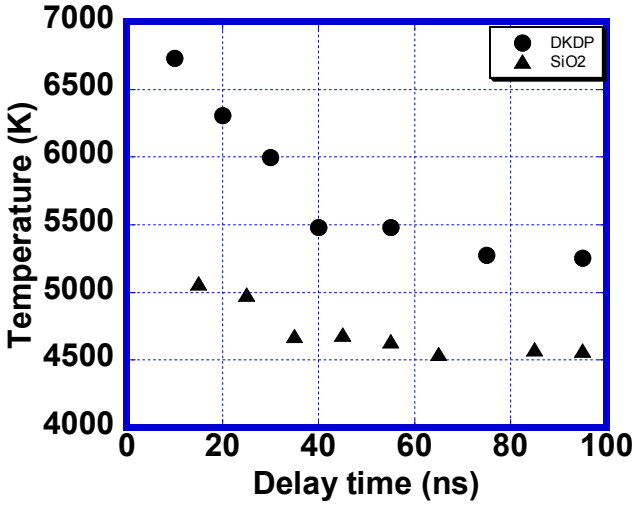


Figure 3

The temperatures extracted from the curve fits of the spectra to equation 1 are plotted as a function of delay time after the pulse.

## DISCUSSION

In a previous work**,** it was observed that the emission from another transparent dielectric (NaCl) was only blackbody at times on the order of 10 μs.[8] In the time interval between the pulse and 1 μs after, the spectra was observed to be non-thermal with a strong blue component. This is entirely consistent with our observations, when the differences in instrumentation are considered. In the previous work the integration times were on the order of microseconds and the spectral window was between 450 nm and 900 nm, in contrast to our 10-ns integration times and 200 to 500-nm spectral window. Because the majority of the emission spectra is in the UV region and is changing on the nanosecond time scale soon after the pulse, we would expect spectra obtained with the longer integration times to appear non-thermal.

In this work we have demonstrated that the emission, which accompanies laser damage, is blackbody in nature at times as short as 10 ns after the laser pulse. The elevated temperatures observed indicate that a substantial amount of energy was absorbed from the laser fields. Because the surrounding material confines the plasma, its density must be essentially unchanged from the solid state. From the equation of state for fused silica, the pressure at the temperatures observed must be on the order of 150 kbars.[9] This yields an energy density on the order of 10 kJ/cm$^3$. We can estimate from the cooling rate that the ionized region is on the order of 5 μm in size[10]. Although this would make the ionized region several orders of magnitude larger in volume (radius ten times larger) than estimated in other works for small solitary inclusions, it is still consistent as we expect that our observed fireballs are a merger or conglomeration of individual defects.[1]

For a fireball of 5 μm in size the energy density corresponds to absorption of roughly 15 μJ, or 2% of the laser pulse. This level of absorption can occur only if the electron density is on the order of the critical density ($10^{22}$ cm$^{-3}$) so that at least a small region is opaque to the laser radiation. The equilibrium electron density of $10^{17}$ cm$^{-3}$ predicted by the Saha formula (electrons promoted across the band gap thermally) for the temperatures observed is several orders of magnitude too small for effective absorption to occur.[4,10] For this reason the electrons cannot be purely thermal in origin. The majority of free electrons must then be produced by impact ionization and electron avalanche.

The radiation can be thermal only if every photon undergoes at least one collision before leaving the ionized region.[10,11] For the electrons to become thermalized in a 5 μm region the electron density must be sufficiently high to reduce the photon mean free path to the same size scale. The photon mean free path depends strongly on the electron density, which must be near or above the critical density to have sufficient effect upon the mean free path. This means that the electron density must also remain relatively high (within a few orders of the critical density) in order for blackbody radiation to be observed after the termination of the laser pulse.

In addition to the cooling rates, several other differences were observed in the behavior of fused silica and DKDP. The DKDP sample would always produce detectable damage on the initial laser pulse or not at all. At very low energies (which will be discussed in detail in a forthcoming work) even if the DKDP would damage on the first laser pulse the same site would not damage on subsequent radiation. The fused silica by contrast would only rarely damage on an initial pulse regardless of the energy used (within the bounds of this study). However multiple exposures to laser energy at the fluence used in this study would generally cause damage.

The distinct differences in initiation suggest that the initial absorption mechanism may be different in the two materials. The DKDP tendency to damage immediately and at relatively low fluencies suggests the presence of defects in substantial quantity before irradiation begins. Fused silica, however, becomes observably damaged only after multiple exposures. Suggesting that early exposures cause a few preexisting defect sites to first damage and then grow until the damage can be detected. Alternately, previous works have demonstrated that x-ray and UV radiation can introduce defects to transparent dielectrics.[12-14] The initial pulses could themselves be introducing defects into the lattice, which after passing a critical density of defects then damage.

The difference in cooling rates and peak temperatures observed in the two materials would be consistent with a larger ionized region in the fused silica. Although it is not conclusive, this could be a result of the purposed damage mechanisms leading to different sized-fireballs.

## CONCLUSION

Time resolved emission spectra observed during laser damage at time greater than 10 ns is found to be blackbody in nature. By fitting the emission spectra we are able to determine the temperature of the plasma fireball that produced the damage. Considerable differences were observed in DKDP and $SiO_2$, including peak temperatures, cooling rates, and damage initiation behavior. The blackbody nature and high temperature of the ionized region indicates electron densities near or above the critical density are reached during laser irradiation and persist for tens of nanoseconds.

## ACKNOWLEDGMENTS

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